Quantum critical phase in $BaVS_3$

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We study the high-pressure metallic phase of high-purity single crystals of $BaVS_3$ by measuring the temperature, pressure, and magnetic field dependence of the resistivity. Above the critical pressure of $p_{\rm cr} = 1.97$GPa an extended non-Fermi liquid $p - T$ regime emerges with resistivity exponent $1.5 \leq n < 2$, crossing over to a FL only around $p = 2.7$GPa. A hysteretic feature indicates that close to the insulator–metal boundary, the system is magnetically ordered. Our findings reveal a close analogy between the extended partially ordered NFL state of non-conventional itinerant magnets and the corresponding state of $BaVS_3$.

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Under atmospheric pressure the linear chain compound $BaVS_3$ undergoes a series of phase transitions [1]. At $T_s = 250$K a hexagonal-to-orthorhombic transition takes place, at $T_{\rm MI} = 70$K a metal-insulator transition (MIT) occurs, which is accompanied by the doubling of the unit cell along the Vanadium chains (c-axis). Finally there is a magnetic transition at $T_X = 30$K whose main aspect is the onset of an incommensurate 3-dimensional magnetic order with a long-period structure along the c-axis [2, 3, 4, 5]. The $T < T_X$ phase was observed to break time reversal invariance, while the other two transitions appear as purely structural [2, 4]. The MIT is broadly speaking a commensurate Peierls transition [6, 7] where the development of a bond/charge order wave [8] opens a gap in the Q1d d-band [9, 10]. Although this transition has a major influence on the susceptibility, and its anisotropy, the corresponding magnetic correlations are not finally settled. Only magnetic quasi-LRO [5] has been observed so far. If any kind of magnetic order occurs below $T_{\rm MI}$ the T_X-transition is its merger with magnetic LRO observed at low temperatures [5, 10].

A phase with incipient LRO should be particularly sensitive to a change of control parameters, including pressure and magnetic field. In contrast, a phase with well-developed order may be less sensitive to such changes [10]. This is evident from the pressure dependence of the phase diagram in Fig 2c: $T_{\rm MI}(p)$ decreases fast with increasing pressure, and the insulating phase is vanishing at a critical pressure $p_{\rm cr} \approx 2$GPa [11]. The transition temperature of the magnetic ordering, $T_X(p)$, is p-independent at least up to ~ 0.7GPa [12], admitting the possibility that the MIT and the magnetic transition merge slightly below 2GPa. Indeed, our recent high-pressure magnetoresistivity studies indicated that in the vicinity of the critical pressure the structural transition and the magnetic ordering phenomenon combine, and in presence of magnetic field a complex order is sought by a hierarchy of very slow relaxation processes [13].

When the insulating phase is completely suppressed by pressure signs of non-Fermi liquid (NFL) behavior emerge [11], similar to those observed in heavy fermion systems near to quantum critical points (QCPs). A quantum phase transition may arise whenever the critical temperature $T_{\rm cr}$ of some kind of long range order is suppressed by a control parameter. In heavy fermion systems, the suppressed order usually has antiferromagnetic character, and $T_{\rm cr} \to 0$ gradually as $p \to p_{\rm cr}$ within the metallic phase. The NFL regime is observed at $p_{\rm cr}$ in a limited part of the parameter space; the Fermi liquid behavior returns as the pressure is further increased from $p_{\rm cr}$. The canonical picture of NFL behavior resulting from a nearby QCP would ascribe it to a wedge-shaped region in the $T - p$ plane, in particular, also to $T \to 0$ at $p = p_{\rm cr}$ [14]. The relationship to underlying microscopic models is complicated [10] but let us note that $BaVS_3$ shares a basic feature with several other NFL systems: the coexistence of wide-band and narrow-band states near the Fermi level [9, 15].

In this paper we investigate the non-Fermi liquid behavior in $BaVS_3$ by varying the control parameters p and B. We show that in this compound the suppression of the combined magnetic and structural order in a quantum phase transition is unlike the simpler QCP phenomena studied previously in heavy fermion systems. Our results reveal NFL behavior not only at the critical pressure, but rather in an extended range of pressure above $p_{\rm cr}$, at sufficiently low temperatures and magnetic fields. The large area covered by the NFL state implies that it does not result from a relatively distant QCP, but rather it is a critical phase as suggested for certain nearly/weakly ferromagnetic systems [16, 17]. Some relevant features of this critical phase is also revealed by high-field magnetotransport measurements.

In an effort to accurately determine the intrinsic low energy properties of $BaVS_3$, special care was taken to use high-quality single crystals. After obtaining crystals by

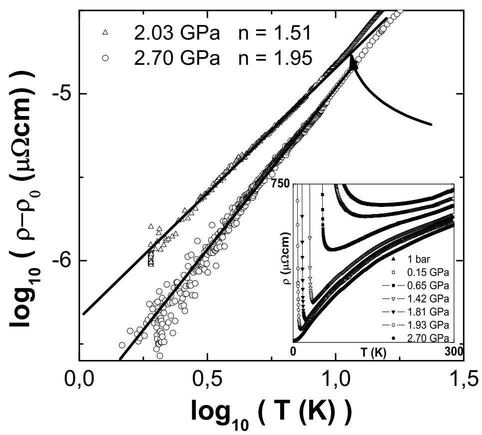

FIG. 1: The low temperature (up to 40 K) part of the resistivity at 2.03 GPa, presented in the log-log plot, reveals the power-law dependence below 15 K. Position of the shoulder (15 K) is indicated by the arrow. The power-law fits (indicated by the straight lines) give the T^n coefficients $n = 1.5$ and $n = 2$ at pressures of 2.03 and 2.7 GPa respectively. Inset: Temperature dependence of resistivity for various pressures showing the suppression of T_{MI}

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the previously established Tellurium flux method [18], a careful characterization was carried out. The single crystals were selected on the basis of the following criteria: i) resistivity measurements at ambient pressure were required to exhibit metallic behavior at high temperatures with a well-defined change of the slope at T_S, a sharp MI transition and no sign of saturation of resistivity in the insulating phase; ii) the resistivity measured at 2 GPa (i.e. at the lowest pressure where the metallic phase extends to whole temperature range) needed to display a high residual resistivity ratio $\rho_{\rm RT}/\rho_{T=2{\rm K}} \sim 50$; iii) magnetic susceptibility anisotropy was required to exhibit both low temperature transitions ($T_{\rm MI}$ and T_X clearly and sharply and no Curie tail at temperatures below 10 K. Samples with a typical dimensions of 2 x 0.2 x 0.2 mm^3 were mounted into a nonmagnetic self-clamped pressure cell where kerosene was used for the pressure transmitting medium. The pressure was monitored in situ using a calibrated InSb pressure gauge. This experimental setup enables resistivity and magnetoresistance measurements over a wide range of temperatures (1.5 to 300 K), pressures (up to 3 GPa) and magnetic fields (up to 12.7 T). The resistivity is measured along the chain direction and the applied magnetic field is perpendicular to the chains. The sweeping rate of the magnetic field was kept slow enough to avoid the heating due to eddy currents.

Figure 1 shows the temperature dependence of the resistivity measured at various pressures. In accordance with previous studies [11] the MIT is suppressed at $\approx$ 2GPa. In the following, we investigate exclusively the high-pressure metallic phase above the critical pressure of $p_{\rm cr} = 2\pm0.02$GPa, where the low temperature resistivity follows a fractional power law behavior with exponent $1.5 \leq n \leq 2$. This customary signature of non-Fermi liquid behavior extends over a broad temperature range (up to 15 K), and a wide pressure interval. Figure 1 displays the two limiting cases. At $p = 2.03$GPa $n = 1.51 \pm 0.01$, the behavior expected of nearly antiferromagnetic systems. At $p = 2.7$GPa $n = 1.95 \pm 0.05$ indicating that Fermi liquid behavior is essentially restored.

To map out the entire intervening regime, the low-temperature resistivity is fitted to

$$\rho(T,p) = \rho_0(p) + A(p)T^{n(p)}$$

with ρ_0, A and n as free parameters. The results are

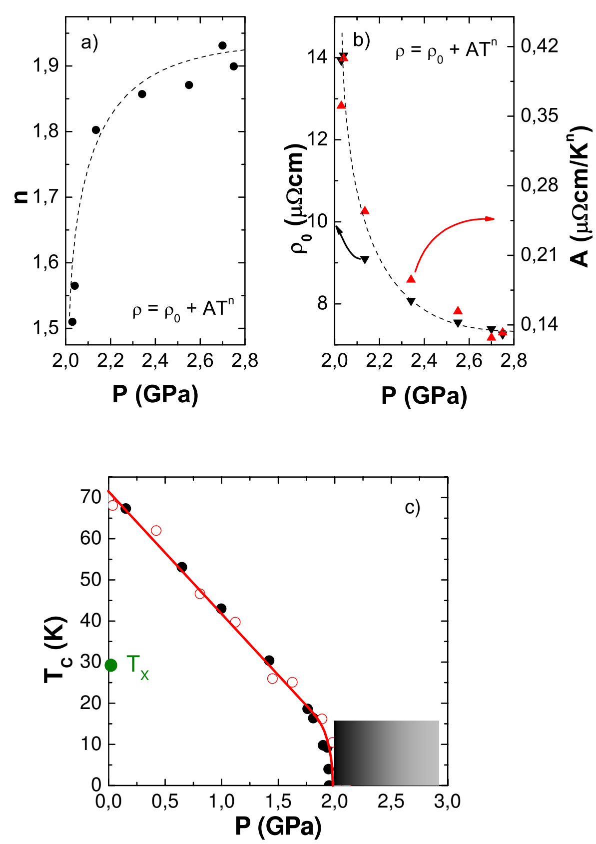

FIG. 2: (Color online) Pressure dependence of the a) resistivity exponent n (the dashed line is to guide the eye), b) Pressure dependence of the prefactor A (red squares) and the "residual" resistivity ρ_0 (black circles) as the results of the fits $\rho = \rho_0 + AT^n$ in the temperature range $1.7 < T < 15$ K. c) The temperature-pressure phase diagram of $BaVS_3$. The MI phase boundary is marked by the red line. The gray regions are related to crossover form NFL to FL, the darkest gray symbolizing n =1.5 and the lightest n = 2.

shown in Fig. 2. The exponent rises continuously from $n \approx 1.5$ to the Fermi liquid value $n = 2$ as p is increased from $p_{\rm cr}$ to $p = 2.7$Gpa. This power-law behavior with gradually changing exponent extends over the grey region displayed in Fig. 2 The pressure dependence of the pre-factor A and the "residual resistivity" ρ_0 is complementary to that of the exponent and both of them seem to diverge as the pressure approaches $p_{\rm cr}$.

Plotting the pressure dependence of A and n is standard in the discussion of NFL systems [19]. However, as shown above, in order to get reasonable fits one must allow the pressure dependence of ρ_0, as well [10]. The result is shown in Fig. 2b. Lowering the pressure towards $p_{\rm cr}$, ρ_0 doubles to $\rho_0(p = 2.03\text{GPa}) = 14\mu\Omega$cm, i.e., the mean free path has been halved to $l \approx 160\mathring{A}$. Since extrinsic defects might not appear due to relaxing the pressure, we ascribe the increment of ρ_0 to the interplay of the anomalous electron correlations with impurity scattering. The scaling of ρ_0, A and n with pressure in Fig. 2 shows that the same intrinsic physics is reflected in those parameters.

In a variety of systems, a regime of NFL behavior was ascribed to strong fluctuations caused by the nearness of a quantum-critical point, with the corollary that Fermi liquid behavior returns gradually as the system is shifted away from the QCP, either with changing pressure or composition, or with applying a magnetic field to suppress spin fluctuations. In the case of $BaVS_3$, the only obvious QCP [20] occurs when the critical value of the control parameter $p_{\rm cr}$ is approached from the metallic side, and the charge gap is suppressed. However, the extension of the incommensurate magnetic order on the insulating side of the QCP to its metallic side may have an influence on the NFL phenomenon.

The exponent $n = 1.5$ is the canonical value expected at the QCP of a nearly antiferromagnetic system [14]. Samples selected by the rigorous criteria described above do not include any with $n < 1.5$. These samples are the most pure $BaVS_3$ crystals, which suggests that the disorder is intrinsic. At the border of the NFL regime $p = 2.7$GPa the residual resistance of $\rho_0 = 7.25\mu\Omega$cm corresponds to a mean free path of 130 V-V interatomic distances, while the system is known as bad-metal with mean free path comparable to the lattice constant at room temperature (or above). Another indication of the good sample quality is that the Fermi liquid form of the electron–electron scattering contribution $\Delta\rho = AT^2$ holds up to approximately 15 K. The $A(p = 2.7\text{GPa}) = 0.14\mu\Omega\text{cm/K}^2$ value on the Kadowaki–Woods plot would mean that high-pressure $BaVS_3$ is analogous to the moderately heavy fermion d-electron system with an expected $\gamma \sim 100(\text{mJ/mole K}^2)$.

Next we investigate a different aspect of the low-temperature metallic phase. At ambient pressure $BaVS_3$ breaks, in succession, point group, translational, and time reversal symmetries, so the question emerges

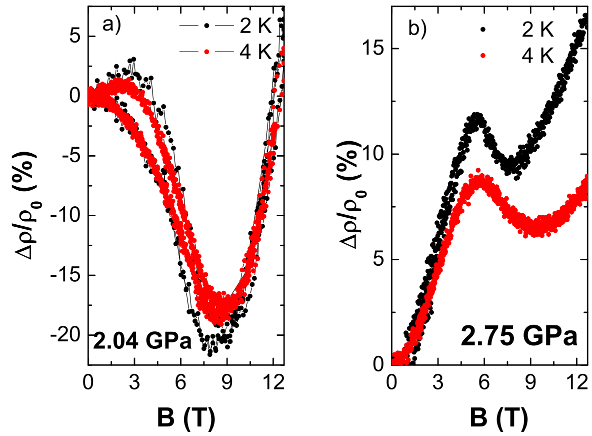

FIG. 3: (Color online) Isothermal magnetoresistance curve recorded at different temperatures and pressures. (a) The 2 and 4 K curves at 2.04 GPa. (b) Curves taken at 2.75 GPa for several temperatures. The cyan arrow indicates the position of the dip for various temperatures.

whether high-p $BaVS_3$ would not undergo some kind of ordering transition. We attempted to find the corresponding resistivity anomaly. At $p = 2.03$GPa the best candidate seems to be a weak shoulder around $T_g \approx$15-20K, which separates the low-temperature NFL regime from the higher-T metallic regime in which ρ rises monotonically from 63 $\mu\Omega cm$ at 20K to $580\mu\Omega cm$ at room temperature. This shoulder resembles (but is not as strong as) the anomaly signaling the onset of ferromagnetic order in $BaVSe_3$ [21]. We also know from studies of the near-critical regime on the insulating side that 15-20K is a significant range of temperature where magnetic degrees of freedom become observable [13]. This motivated extending our magnetoresistivity studies to the high-pressure regime.

Figure 3a shows the magnetic field dependence of the low-temperature resistivity at $p = 2.04$GPa, i.e., slightly above the critical pressure. The striking feature is the existence of hysteresis which proves that metallic $BaVS_3$ possesses some kind of magnetic order at these temperatures. As the only notable feature of the temperature dependence of the resistivity is the shoulder at T_g, we guess that this is the ordering temperature. The nature of the order may be either structural distortion (such as the low-pressure tetramerization) or magnetic order. We exclude the former since the energetic motivation of tetramerization would be the opening of spin and charge gaps. We also note that the MIT has a structural [3] and resistivity [1] precursor and it is known that the resistivity precursor is suppressed at the same pressure where the ground state becomes metallic [11]. Thus the $T < T_g$ high-pressure metallic phase of $BaVS_3$ should be isostructural with the ambient-pressure $T_{\rm MI} < T < T_S$ phase,

and it is magnetically ordered.

In our interpretation the quantum phase transition at $p = p_{\rm cr}$ is a magnetic-insulator-to-magnetic-metal transition. The high-pressure magnetic structure is unknown. It cannot be exactly the same as at $p < p_{\rm cr}$, as it exists on a different crystalline background, and spin–orbit coupling is relevant [1]. Still, it is likely to be similar to the ambient-pressure low temperature magnetic state [5] in the sense of having some long period [23]. This allows to interpret the B-cycle hysteresis shown in Figure 3a by invoking our previous argument for similar phenomena in the insulating phase [13]. We envisage that the effect of changing the pressure and other thermodynamical parameters, like T and B, is changing $\mathbf{Q}$, the ordering vector of the magnetic structure, or its aligning (casused by the relativistic spin–orbit coupling which is manifest in the magnetic anisotropy [1]) with the lattice. Hysteresis should remain observable as long as there is a magnetic order with finite period. Since hysteresis is definitely absent at $p = 2.75$GPa (Fig. 3b), where Fermi liquid behavior is restored, we suggest the existence of a magnetic phase at $T < T_g$ in the $p - T$ domain with unambiguously NFL behavior.

NFL behavior is not typical of structurally ordered metallic magnets. This shows that the magnetic order below the $T_g(p)$ line is not necessarily the conventional long-range order of either a SDW/AFM or a FM system. We rather propose a state analogous to the partially ordered extended NFL regime of MnSi [22]. It has been claimed that FL theory or its straightforward extension fail to describe some ordered phases in nearly/weakly FM materials [16, 17]. Lacking direct evidence, we would rather refrain from stating that the similarity would go as far as high-pressure $BaVS_3$ being actually a partially ordered FM [23]. There may exist generalizations of the novel metallic phase discussed for MnSi to the systems with coupled magnetic and bond/charge orders.

In conclusion, clean high-quality $BaVS_3$ possesses an extended NFL regime which at the same time has some kind of magnetic order. The novel state of magnetism in good samples with mean free paths $l \sim O(100\mathring{A})$ is characterized by diverging low-temperature electron–electron scattering evident in $\Delta\rho \propto T^n$, with exponent $1.5 \leq n < 2$, and hysteresis phenomena indicating magnetic order.

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69, 3068 (2000)), and $Ba_{1-x}Sr_xVS_3$ (A. Gauzzi et al, cond-mat/0601286). However, unlike those systems $BaVS_3$ is a particularly perfect and clean system, a feature which was also emphasized in the analogous studies on MnSi.